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### Remarks

Further and favorable reconsideration is respectfully requested in view of the foregoing amendments and following remarks.

Thus, the claims have been amended along the lines suggested by the Examiner in items (1) and (2) on page 2 of the Office Action to improve the form and clarity of the claims.

Claims 30 and 36 have been amended in response to the rejection of these claims under the second paragraph of 35 U.S.C. §112, rendering this rejection moot.

In this regard, claim 30 has been amended to specify a weight basis for the amount of terpenic alcohols. A weight basis would be apparent to one of ordinary skill in the art because this is the quantity accessible from the analysis of complex mixtures such as pine oils by gas phase chromatography (GPC), as supported by the enclosed copy of Kirk-Othmer Encyclopaedia of Chemical Technology, Vol. 22, pages 747-749.

Claim 36 has been amended along the lines suggested by the Examiner, and also recites at least one surfactant, which is supported by the disclosure at page 4, line 29 of the specification.

Further amendments have been introduced into the claims which result in mooting all of the prior art rejections set forth by the Examiner.

Thus, referring to the rejection of claims 20, 26, 31-34, 37 and 38 under 35 U.S.C. §102(b) as being anticipated by Clark et al., each of the independent claims subject to this rejection, i.e. claims 20, 37 and 38, has been amended to recite at least one monoterpene. These claims as amended thus incorporate the limitation of claim 25 (which has thus been cancelled) which the Examiner indicates would be allowable if rewritten in independent form (page 8 of the Office Action).

In rejecting claim 37 under 35 U.S.C. §102(e) as being anticipated by Kierzkowski et al., the Examiner notes that this claim does not actually recite that the copper must be suspended. Claim 37 has been amended to refer to the copper compound "in suspension . . . ."

Claims 21-23, which have been rejected under 35 U.S.C. §103(a) as being unpatentable over Clark et al. in view of Farm Chemicals Handbook '98, are all composition claims. Claim 20, on which claims 21-23 depend, has been amended as indicated above, to incorporate the subject matter of claim 25 which the Examiner indicates would be allowable if rewritten in independent form.

Thus, it is apparent that all of the prior art rejections have been rendered moot.

Therefore, in view of the foregoing amendments and remarks, it is submitted that each of the grounds of objection and rejection set forth by the Examiner has been overcome, and that the application is in condition for allowance. Such allowance is solicited.

Respectfully submitted,

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Other methods for the synthesis of camphor exist but are not commercially important. The 1981 price of the synthetic product is ca \$4-6/kg (53).

**Uses.** Camphor is used chiefly as an ingredient in pharmaceutical preparations, as a moth repellent (see Repellents in Supplement Volume), a religious incense, and an odorant-flavorant in a variety of consumer products.

**Pine Oil.** Except for acetylation and hydrogenation of  $\alpha$ -terpineol-rich fractions isolated from it, pine oil undergoes no important commercial reactions.

**Manufacture and Price.** There are three types of pine oil: steam-distilled, sulfate, and synthetic. Steam-distilled pine oil, a product of the wood naval-stores industry, is obtained by distillation of the extract from aged pine stumps in the southeastern United States. Sulfate pine oil is obtained by the fractionation of crude sulfate turpentine. Synthetic pine oil is produced by the acid-catalyzed hydration of pinene, usually  $\alpha$ -pinene, followed by distillation. Mineral acids are commonly used in strengths of 20-40 wt % and at ca 30-50°C. The hydration is carried out to produce a maximum of alcohols and a minimum of hydrocarbons and cineoles by controlling the reaction variables of time, temperature, and acid strength. The reaction proceeds through carbonium ion mechanisms. Formation of the monocyclic products is depicted below. Some of the reactions are reversible under the acid reaction conditions.

The chief oxygenated components of pine oils are  $\alpha$ -terpineol,  $\beta$ -terpineol [138-87-4],  $\gamma$ -terpineol,  $\alpha$ -fenchol, borneol, isoborneol, camphor, terpinen-1-ol, terpinen-4-ol, dihydroterpineol [498-81-7], methyl chavicol [140-67-0], anethole [104-46-1], and 1,4- and 1,8-cineole. Not all of these components are present in all pine oils, but all pine oils contain  $\alpha$ -terpineol as the main oxygenated component. In addition to the oxygenated components, all pine oils contain amounts of *p*-menthadienes, the most predominant ones being limonene (dipentene), terpinolene, and  $\alpha$ -terpinene, with smaller amounts of 2,4(8)-*p*-menthadiene and  $\gamma$ -terpinene; small amounts of pinene and cymene may also be present.

Many grades of pine oil are available commercially and differ according to the source, the efficiency and type of distillation, and the way the various distillation cuts are combined. A high grade synthetic pine oil has the composition shown in Table 4, as determined by capillary gas chromatography.

Synthetic pine oils usually have higher terpinolene contents than the other pine oils, and only trace amounts of camphor are present. They contain no appreciable amounts of sesquiterpenes and do not contain anethole and methyl chavicol, which are usually in the other types of pine oil. Steam-distilled pine oil is relatively high in borneol and fenchol. Commercial pine oils are specified by physical properties and by alcohol content, as determined by either ASTM D 802-57 or gas chromatography. A pine oil, such as described above, typically has the properties listed in Table 5.

Lower grades of  $\alpha$ -terpineol for odorant use can be obtained by distillation from pine oil, but these always contain small amounts of borneol,  $\gamma$ -terpineol, or both. High grade perfumery  $\alpha$ -terpineol is made by partial dehydration of terpin hydrate [2451-01-6] under weakly acidic conditions (165). A substantial portion of  $\alpha$ -terpineol produced is converted to its acetate [80-26-2] or dihydro- $\alpha$ -terpineol and its acetate which are also widely used in perfumery. U.S. production of synthetic pine oil in 1980 amounted to 20,000 t at an average price of \$1.20/kg (53).

**Uses.** By far the largest use of pine oil is in the manufacture of cleaners and disinfectants; it is effective against gram-negative enterobacteria but not against gram-positive organisms. Lower grades, eg, pine-oil sulfate, are used as frothers in separating



Table 4. Composition of Synthetic Pine Oil

Alcohols	Wt %	Others	Wt %
$\alpha$ - and $\gamma$ -terpineol	67.0	terpinolene	6.7
terpinene-1-ol	3.9	limonene	2.1
<i>cis</i> - $\beta$ -terpineol	2.8	<i>p</i> -2,4(8)-menthadiene	1.7
isoborneol	2.2	$\alpha$ -terpinene	1.1
borneol	2.1	1,8-cineole	0.8
terpinene-4-ol	2.0	camphor	0.7
fenchol	1.9	1,4-cineole	0.6
<i>trans</i> - $\beta$ -terpineol	1.3	$\gamma$ -terpinene	0.3
trace alcohols	2.6	<i>p</i> -cymene	0.3

Table 5. Physical Properties of Synthetic Pine Oil

Property	Value
specific gravity (15.5/15.5°C), min	0.930
terpene alcohols <sup>a</sup> , wt %, min	85
moisture, wt %, max	0.5
color (APHA)	10
refractive index, $n_D^{20}$	1.479–1.489
acid number	<1
flash point, °C	76
Kauri butanol value	>500
distillation range, %	
<195°C, max	5.0
<230°C, min	95.0

<sup>a</sup> As measured by gas chromatography.

minerals by flotation. Some of the attributes of pine oil that make it useful in the manufacture of glues and adhesives (qv) are its dispersive and solvent power, its surface activity, and its preservation and plasticizing properties. Another use of pine oil is in the textile industry as a penetrant, dispersing agent, wetting agent, and inhibitor of bacterial growth in wet-processing of cotton, silk, rayon, and woolen goods. It also is used in the protective-coatings, agricultural-chemicals, packing-house, and leather industries (see Disinfectants).

**Terpene Resins.** Large quantities of terpene hydrocarbons are used in the manufacture of terpene resins. The most commonly used terpenes are  $\beta$ -pinene,  $\alpha$ -pinene, limonene, or a combination of these. Other terpenes have been used, but to much less extent. The cationic polymerization is typically accomplished by reaction in a solvent, eg, toluene, and catalyzed by a Lewis acid,  $AlCl_3$  being the most commonly used. The resins are thermoplastic and generally of light color (Gardner 2–5). Terpenes have also been copolymerized with a variety of nonterpene monomers. The largest use of terpene resins is in hot-melt and pressure-sensitive adhesives, although other significant uses exist.